Supplementary Materials for 'Comparing the molecular and global rheology of a fluid under high pressures'

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S.1 Sensitivity of the Barus equation to fitting range

While Roelands model and the Hybrid model describe our experimental results very well, they both have their limitations. The former can only be used before the faster than exponential region is reached. There are other more complex models, such as the improved-Yasutomi model[1] and the full Hybrid model, which are capable of fitting data across a very large pressure range. However, they require constants, such as the McEwen parameter and the Fragility parameter[2], that can be difficult to calculate or measure and are usually found by regression. Single exponential models are attractive because of their simplicity. However, the free single exponential fit and the Barus equation fit perform poorly at low pressure and high-pressure conditions respectively. This is because of the changing compressibility of the fluids with pressure. To assess the predictive power of the Barus equation in this work, α are obtained from low-pressure viscosity data, by limiting the maximum pressure to be considered for fitting purpose. The resulting equation is then used to predict high-pressure viscosity. Predicted and experimental results are compared. Varying the range of pressure for fitting the Barus equation affects both the value of α and the goodness of fit R^2, as shown in Figure S1. As the pressure range increases, α first decreases then plateaus. This shows that α obtained from data at very low pressures only do not have practical use as it is too sensitive to pressure change. At pressure limit of 100 - 250 MPa (within the blue solid ellipse), the drop in α has slowed down and the α may be used as an approximation of α_0 value. If the pressure limit is now set to 250 - 450 MPa (green dash ellipse), α is fairly constant. While this α cannot be used as α_0, this can be used as α_p up to the specified pressure limit. Some extrapolation can be undertaken but care must be taken. Increasing the pressure limit further increases R^2 in the expense of poor fit at the lower pressure regions. Hence should not be used if lower pressure viscosity is of interest.

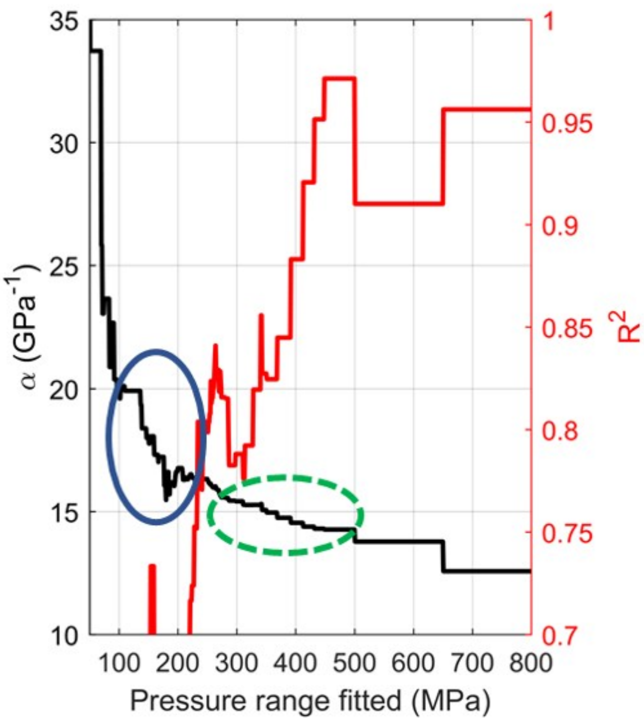

Figure S1: Effect of maximum fitting pressure on α within the Barus equation. It shows usable range where data up to 75-100 MPa (Blue solid region) gives a consistent usable α_0 then next 200-450 (Green dash region) MPa reliable for an α value.

The effect of pressure on pressure-viscosity coefficient is. As an example, α estimated from fluid film thickness measurements[3] using an EHD contact is based on the inlet pressure (see Figure 1 for the definition of the inlet) as the thickness of the film, h, is predominately controlled by the inlet conditions. Hence the pressure-viscosity coefficient obtained approximates α at the inlet pressure. On the other hand, pressure-viscosity coefficient taken from friction measurements with varying load may be more representative of α at a pressure inside the contact area. These α values can be very different, and care must be taken when choosing how to measure and define pressure-viscosity coefficients.

S.2 Rheological Data of PAO 8 at 22 ℃

Table S1 shows the raw data for PAO 8, obtained through private communication, measured in a high pressure rehometer under Newtonian conditions[4]. Further rheolgical data for various PAOs at high pressure can be found in the work of Bair and Flores-Torres[5].

Table S1: Rheological data for PAO 8 at high pressure at under Newtonian conditions[4].

p (MPa)	η (Pa.s)
0	0.0871
25	0.142
50	0.231
100	0.519
150	1.04
250	3.84
350	12.6
500	64.4
650	298
800	1310
900	4240

S.3 Deviation plot of η^* & η^A against η

Figure S2 Shows η^* & η^A plotted against η at the same pressure. See Table 1 for definitions. η is matched to the experimental pressures, by fitting η against p using equation 23. Dashed blue line for clarity, showing where the two data sets are equal.

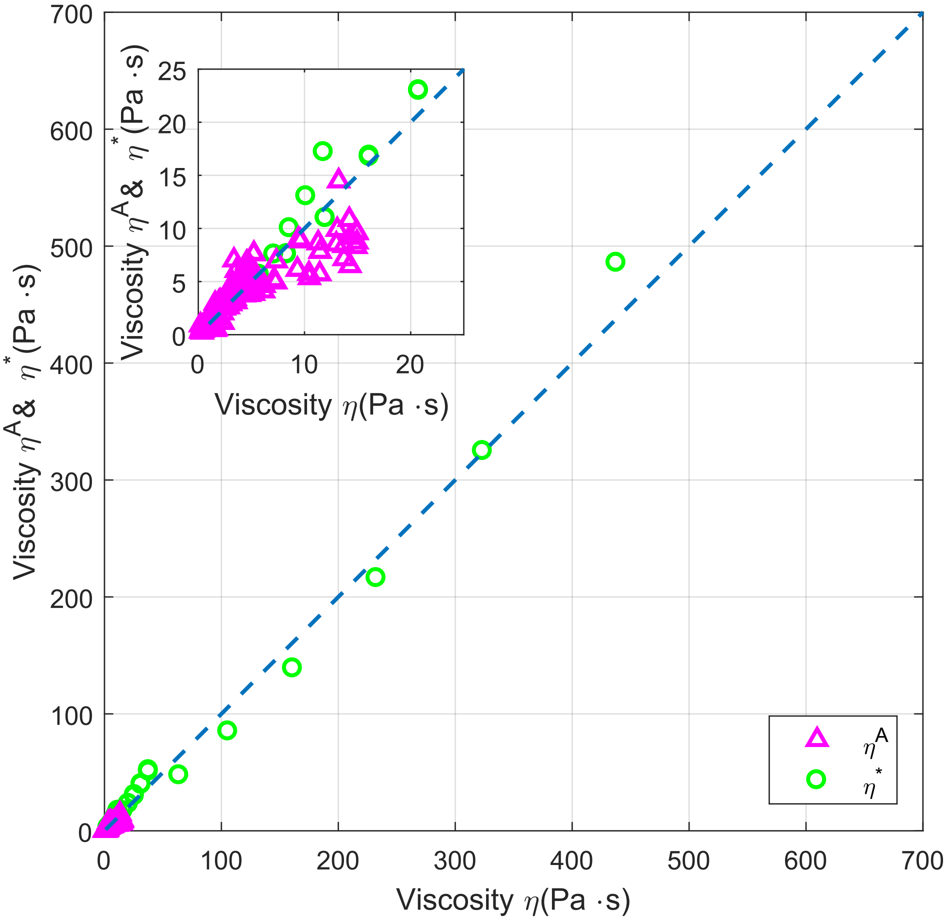

Figure S2: Shows η^* & η^A plotted against η at the same pressure. See Table 1 for definitions. η is matched to the experimental pressures, by fitting η against p using equation 24. Dashed blue line for clarity, showing where the two data sets are equal.

References

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3 H. van Leeuwen, *Proc. Inst. Mech. Eng., Part J J. Eng. Tribol.*, 2011, **225(6)**, 449–464.

4 S. S. Bair, 2018.

5 S. Bair and S. Flores-Torres, *J. Tribol.*, 2018, **141**, 021802.